

CLAIMS

1. A crystalline mesoporous silica material being obtained by assembly of nanometer size building units having zeolite framework, said crystalline mesoporous silica material having two or more levels of porosity and structural order, and wherein the internal structure of said nanometer size building units does not give rise to Bragg type diffraction in a powder X-ray diffraction pattern of said crystalline mesoporous silica material.
2. A crystalline mesoporous silica material according to claim 1, wherein said building units are nanoslabs of substantially uniform size having the Silicalite-1 zeolite framework.
3. A crystalline mesoporous silica material according to claim 2, wherein said nanoslabs are generated by tetrapropylammonium-ion mediation.
4. A crystalline mesoporous silica material according to claim 2 or claim 3, wherein said nanoslabs are linked through their corners, edges or faces following patterns imposed by interaction with a cationic surfactant or triblock copolymer molecule.
5. A crystalline mesoporous silica material according to any of claims 2 to 4, wherein said two or more levels of porosity comprise at least a microporosity inside said nanoslabs and at least a mesoporosity between said nanoslabs.
6. A crystalline mesoporous silica material according to claim 4, wherein said cationic surfactant is cetyltrimethylammonium bromide.
7. A crystalline mesoporous silica material according to claim 4, wherein said triblock copolymer is a Pluronic P123 triblock copolymer  $\text{EO}_{20} \text{PO}_{70} \text{EO}_{20}$ .
8. A crystalline mesoporous silica material according to claim 4 or claim 6, being Zeotile 1, the powder X-ray diffraction pattern and high resolution

electron microscopy micrograph thereof being substantially as shown in figure 1.

9. A crystalline mesoporous silica material according to claim 8, wherein nanoslabs are forced into face-sharing double units with a size ranging  
5 from 2 to 4 nm and linked in a hexagonal symmetry pattern.
10. A crystalline mesoporous silica material according to claim 4 or claim 7, being Zeotile 4, the high resolution electron microscopy micrograph thereof being substantially as shown in figure 3.
11. A crystalline mesoporous silica material according to claim 10, being  
10 obtained from nanoslabs with a size ranging from 1.3 to 8 nm.
12. A crystalline mesoporous silica material according to claim 10 or claim 11, being obtained from nanoslabs with a size ranging from 4 to 8 nm.
13. A crystalline mesoporous silica material according to any of claims 10 to 12, being obtained from stapled nanoslabs arranged in a hexagonal  
15 appearing tile.
14. Use of a crystalline mesoporous silica material according to any of claims 1 to 13 for the fixation of biologically active species.
15. Use of a crystalline mesoporous silica material according to any of claims 1 to 13 as an electro-optical or dielectric material or for catalysis,  
20 molecular separation or adsorption of metal ions.
16. Use of a cationic surfactant or a triblock copolymer molecule for the assembly of nanoslabs of substantially uniform size having the Silicalite-1 zeolite framework.
17. Use according to claim 16, wherein said assembly proceeds while  
25 imparting a structural organisation to said nanoslabs.

18. Use according to claim 16 or claim 17, wherein said nanoslabs are linked through their corners, edges or faces following patterns imposed by interaction with said cationic surfactant or triblock copolymer molecule.
19. Use according to claim 16, wherein said cationic surfactant is cetyltrimethylammonium bromide.
20. Use according to claim 16, wherein said triblock copolymer is a Pluronic P123 triblock copolymer  $EO_{20} PO_{70} EO_{20}$ .
21. A process for making a crystalline mesoporous silica material according to any of claims 2 to 4, comprising the steps of (a) generating nanoslabs having zeolite framework by tetrapropylammonium-ion mediation and (b) assembling said nanoslabs through interaction with a cationic surfactant or triblock copolymer molecule.
22. A process according to claim 21, wherein said cationic surfactant is cetyltrimethylammonium bromide.
23. A process according to claim 21, wherein said triblock copolymer is a Pluronic P123 triblock copolymer  $EO_{20} PO_{70} EO_{20}$ .
24. A process according to any of claims 21 to 23, further comprising the step of removing said tetrapropylammonium-ion and said cationic surfactant or triblock copolymer molecule.
25. A process according to claim 24, wherein removing said tetrapropylammonium-ion and said cationic surfactant or triblock copolymer molecule is effected through oxidation or calcination.
26. A crystalline mesoporous silica material according to claim 1, wherein said absence of Bragg type diffraction from nanometer size building units manifests itself by the absence of peaks in a powder X-ray diffraction pattern at interplanar spacings below about 1.5 nm.

27. A crystalline mesoporous silica material according to claim 1 or claim 26, wherein said powder X-ray diffraction pattern is performed after removal of said tetrapropylammonium-ion and said cationic surfactant or triblock copolymer molecule.
- 5 28. A substantially crystalline mesoporous oxide based material being obtained by assembly of nanometer size building units having zeolite framework, wherein said assembly proceeds in the presence of one or more amphiphilic non-anionic surfactants, wherein said substantially crystalline mesoporous oxide based material has two or more levels of porosity and structural order, and wherein the internal structure of said  
10 nanometer size building units does not give rise to Bragg type diffraction in a powder X-ray diffraction pattern of said substantially crystalline mesoporous oxide based material.
29. A substantially crystalline mesoporous oxide based material according to  
15 claim 28, wherein said absence of Bragg type diffraction from nanometer size building units manifests itself by the absence of peaks in a powder X-ray diffraction pattern at interplanar spacings below about 1.5 nm and/or  
( ) at angles of diffraction above 3 degrees.
30. A substantially crystalline mesoporous oxide based material according to  
20 claim 28 or claim 29, wherein said powder X-ray diffraction pattern is performed after removal of said one or more amphiphilic non-anionic surfactants.
31. A substantially crystalline mesoporous oxide based material according to  
25 any of claims 28 to 30, wherein said oxide based material comprises one or more oxides selected from the group consisting of silica, germanium oxide and metallic oxides.
32. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 31, wherein said oxide based material comprises one or more metallic oxides selected from the group consisting of alumina,

titania, zirconia, ceria, manganese oxide, niobium oxide, tantalum oxide, tungsten oxide, tin oxide, gallium oxide, iron oxide, and hafnium oxide.

33. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 32, wherein the size of said building units ranges from about 1 to 8 nm.

34. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 33, having one or more types of mesopores each with an average size ranging from about 2 to 15 nm.

35. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 34, having one or more types of mesopores each having a narrow pore size distribution, as calculated by the Barrett-Joyner-Halenda analysis from the nitrogen adsorption/desorption isotherm of said mesoporous oxide based material.

36. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 35, wherein said nanometer size building units are nanoslabs of substantially uniform size having a zeolite framework.

37. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 36, wherein said nanometer size building units are nanoslabs of substantially uniform size having the Silicalite-1 zeolite framework.

38. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 37, wherein said nanometer size building units are generated by means of a mediating agent selected from the group consisting of tetraalkylammonium ions, tetraalkylphosphonium ions and gemini (dimeric) tetraalkylammonium ions wherein the alkyl group has from 2 to 4 carbon atoms.

39. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 38, wherein said alkyl group is propyl.

40. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 39, wherein said nanometer size building units are  
5 linked through their corners, edges or faces following patterns imposed by interaction of said nanometer size building units with said one or more amphiphilic non-anionic surfactants.

41. A substantially crystalline mesoporous oxide based material according to claim 40, wherein said one or more amphiphilic non-anionic surfactants  
10 are selected from the group consisting of amphiphilic non-ionic molecules and amphiphilic cationic molecules.

42. A substantially crystalline mesoporous oxide based material according to claim 40 or claim 41, wherein said one or more amphiphilic non-anionic  
15 substances are selected from the group consisting of amphiphilic block copolymers, cationic gemini (dimeric) surfactants and C<sub>12-18</sub> alkyltrimethylammonium halide surfactants.

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43. A substantially crystalline mesoporous oxide based material according to any of claims 40 to 42, wherein said one or more amphiphilic non-anionic  
20 substances is a C<sub>12-18</sub> alkyltrimethylammonium halide surfactant or a cationic gemini (dimeric) surfactant, and wherein interaction proceeds under basic conditions.

44. A substantially crystalline mesoporous oxide based material according to claim 43, wherein interaction proceeds in the presence of one or more salts selected from the group consisting of aluminates, borates and acid  
25 salts of 3d transition metals on the periodic table.

45. A substantially crystalline mesoporous oxide based material according to any of claims 40 to 42, wherein said one or more amphiphilic non-anionic

substances is an amphiphilic block copolymer and wherein interaction proceeds under acidic conditions.

46. A substantially crystalline mesoporous oxide based material according to any of claims 40 to 42, wherein said one or more amphiphilic non-anionic substances is a poly(ethylene oxide)-poly(alkylene oxide)- poly(ethylene oxide) triblock copolymer wherein the alkylene oxide moiety has at least 3 carbon atoms.
47. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 46, wherein said two or more levels of porosity comprise at least a microporosity inside said nanometer size building units and at least a mesoporosity between said nanometer size building units.
48. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 47, wherein said building units are assembled or arranged in a hexagonal pattern.
49. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 47, being obtained from building units with a size ranging from about 1 to 8 nm.
50. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 49, being Zeotile 2, the powder X-ray diffraction pattern and high resolution electron microscopy micrograph thereof being substantially as shown in figure 2.
51. A substantially crystalline mesoporous oxide based material according to any of claims 28 to 50, having mesopore walls with a thickness from about 1 to 4 nm.
52. Use of a substantially crystalline mesoporous oxide based material according to any of claims 28 to 51 for the fixation or immobilisation of a biologically active species.

53. Use according to claim 52, wherein said biologically active species is a poorly soluble therapeutic drug.
54. Use according to claim 52 or claim 53, wherein said poorly soluble therapeutic drug is classifiable as belonging to Class II or Class IV of the Biopharmaceutical Classification System.
55. Use according to any of claims 52 to 54, wherein the size of said biologically active species is suitable for entrapment into the mesopores of said substantially ordered mesoporous oxide based material.
56. Use according to any of claims 52 to 55, wherein the molecular weight of said biologically active species is between about 200 and 1,000.
57. Use according to any of claims 52 to 56, wherein said biologically active species is itraconazole or diazepam.
58. Use of an amphiphilic non-anionic molecule for the assembly of nanometer size building units having zeolite framework.
59. Use according to claim 58, wherein said assembly proceeds while imparting a structural organisation to said nanometer size building units.
60. Use according to claim 58 or claim 59, wherein said nanometer size building units are linked through their corners, edges or faces following patterns imposed by interaction with said amphiphilic non-anionic molecule.
61. Use according to any of claims 58 to 60, wherein said amphiphilic non-anionic molecule is selected from the group consisting of amphiphilic non-ionic molecules and amphiphilic cationic molecules.
62. Use according to any of claims 58 to 61, wherein said amphiphilic non-anionic molecule is selected from the group consisting of amphiphilic



block copolymers, cationic gemini (dimeric) surfactants and C<sub>12-18</sub> alkyltrimethyl-ammonium halide surfactants.

63. Use according to any of claims 58 to 62, wherein said amphiphilic non-anionic molecule is cetyl-trimethylammonium bromide.

5 64. Use according to any of claims 58 to 62, wherein said molecule is a poly(ethylene oxide)-poly(alkylene oxide)-poly(ethylene oxide) triblock copolymer wherein the alkylene oxide moiety has at least 3 carbon atoms.

65. Use according to any of claims 58 to 64, wherein the size of said nanometer size building units ranges from about 1 to 8 nm.

10 66. Use according to any of claims 58 to 65, wherein said nanometer size building units comprise one or more oxides selected from the group consisting of silica, germanium oxide and metallic oxides, and wherein said assembly forms a crystalline mesoporous oxide based material.

15 67. A process for making a substantially crystalline mesoporous oxide based material according to any of claims 28 to 51, comprising the steps of (a) generating nanometer size building units having zeolite framework by means of a mediating agent selected from the group consisting of tetraalkylammonium ions, tetraalkylphosphonium ions and gemini (dimeric) tetraalkylammonium ions wherein the alkyl group has from 2 to 4  
20 carbon atoms, and (b) assembling said nanometer size building units through interaction with one or more amphiphilic non-anionic substances to yield a mesoporous oxide based material.

68. A process according to claim 67, wherein said one or more amphiphilic non-anionic substances are selected from the group consisting of  
25 amphiphilic non-ionic molecules and amphiphilic cationic molecules.

69. A process according to claim 67 or claim 68, wherein said one or more amphiphilic non-anionic substances are selected from the group

consisting of amphiphilic block copolymers, cationic gemini (dimeric) surfactants and C<sub>12-18</sub> alkyltrimethyl-ammonium halide surfactants.

70. A process according to any of claims 67 to 69, wherein said one or more amphiphilic non-anionic substances is a C<sub>12-18</sub> alkyltrimethylammonium halide surfactant or a cationic gemini (dimeric) surfactant, and wherein interaction proceeds under basic conditions.

71. A process according to claim 70, wherein interaction proceeds in the presence of one or more salts selected from the group consisting of aluminates, borates and acid salts of 3d transition metals on the periodic table.

72. A process according to any of claims 67 to 69, wherein said one or more amphiphilic non-anionic substances is an amphiphilic block copolymer and wherein interaction proceeds under acidic conditions.

73. A process according to claim 72, wherein said amphiphilic block copolymer is a poly(ethylene oxide)-poly(alkylene oxide)-poly(ethylene oxide) triblock copolymer wherein the alkylene oxide moiety has at least 3 carbon atoms.

74. A process according to any of claims 67 to 73, further including the step of calcining or solvent extracting the mesoporous oxide based material from step (b) for substantially removing the organic species introduced in steps (a) and (b).

75. A process according to claim 74, wherein removing said organic species introduced in steps (a) and (b) is effected by means of oxidation, calcination or solvent extraction.

76. Use of a substantially ordered mesoporous oxide based material for the fixation or immobilisation of a biologically active species, wherein said ordered oxide based material has one or more levels of porosity and

structural order, provided that when said ordered oxide based material has a single level of porosity and structural order it is obtained in the absence of an alpha-tocopherol polyethylene glycol ester templating biomolecule.

5 77. Use according to claim 76, wherein said biologically active species is a poorly soluble therapeutic drug.

78. Use according to claim 76 or claim 77, wherein said poorly soluble therapeutic drug is classifiable as belonging to Class II or Class IV of the Biopharmaceutical Classification System.

10 79. Use according to any of claims 76 to 78, wherein the size of said biologically active species is suitable for entrapment into the mesopores of said substantially ordered mesoporous oxide based material.

80. Use according to any of claims 76 to 79, wherein the molecular weight of said biologically active species is between about 200 and 1,000.

15 81. Use according to any of claims 76 to 80, wherein said biologically active species is itraconazole or diazepam.

82. Use according to any of claims 76 to 81, wherein said substantially ordered mesoporous oxide based material has two or more levels of porosity and structural order.

20 83. Use according to claim 82, wherein said substantially ordered mesoporous oxide based material having two or more levels of porosity and structural order is obtained by assembly of nanometer size building units having zeolite framework and wherein said assembly proceeds in the presence of one or more amphiphilic non-anionic surfactants.

25 84. Use according to claim 83, wherein said nanometer size building units do not give rise to Bragg type diffraction in a powder X-ray diffraction pattern of said substantially crystalline mesoporous oxide based material.

85. Use according to claim 84, wherein said absence of Bragg type diffraction from nanometer size building units manifests itself by the absence of peaks in a powder X-ray diffraction pattern at interplanar spacings below about 1.5 nm and/or at angles of diffraction above 3 degrees.

5 86. Use according to claim 84 or claim 85, wherein said powder X-ray diffraction pattern is performed after removal of said one or more amphiphilic non-anionic surfactants.

87. A pharmaceutical composition comprising a biologically active species and a substantially ordered mesoporous oxide based material, wherein  
10 said ordered oxide based material has one or more levels of porosity or structural order, provided that when said ordered oxide based material has a single level of porosity and structural order it is obtained in the absence of an alpha-tocopherol polyethylene glycol ester templating biomolecule.

88. A pharmaceutical composition according to claim 87, wherein said  
15 substantially ordered mesoporous oxide based material has two or more levels of porosity and structural order.

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89. A pharmaceutical composition according to claim 88, wherein said substantially ordered mesoporous oxide based material having two or more levels of porosity and structural order is obtained by assembly of  
20 nanometer size building units having zeolite framework.

90. A pharmaceutical composition according to claim 88 or claim 89, wherein said nanometer size building units do not give rise to Bragg type diffraction in a powder X-ray diffraction pattern of said substantially crystalline mesoporous oxide based material.

25 91. A pharmaceutical composition according to any of claims 88 to 90, wherein said mesoporous oxide based material is a material according to any of claims 28 to 51.

92.A pharmaceutical composition according to any of claims 88 to 91, wherein said oxide based material is the silica material named Zeotile 4, the high resolution electron microscopy micrograph thereof being substantially as shown in figure 3.

5 93.A pharmaceutical composition according to any of claims 87 to 92, wherein the size of said biologically active species is suitable for entrapment into the mesopores of said substantially ordered mesoporous oxide based material.

10 94.A pharmaceutical composition according to any of claims 87 to 93, wherein said biologically active species is a poorly soluble therapeutic drug.

95.A pharmaceutical composition according to any of claims 87 to 94, wherein said poorly soluble therapeutic drug is classifiable as belonging to Class II or Class IV of the Biopharmaceutical Classification System.

15 96. A pharmaceutical composition according to any of claims 87 to 95, wherein the molecular weight of said biologically active species is between about 200 and 1,000.

97.A pharmaceutical composition according to any of claims 87 to 96, wherein said poorly soluble therapeutic drug is itraconazole or diazepam.

20 98.A pharmaceutical composition according to any of claims 87 to 97, further comprising one or more pharmaceutically acceptable excipients.

99.A pharmaceutical composition according to any of claims 87 to 98, providing immediate *in vivo* release of said biologically active species.